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Title: Corrosion Research Survey: Corrosion-Surface Roughness Correlation on Carbon and Stainless Steels

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Corrosion Research Survey:

Corrosion-Surface Roughness Correlation on
Cabon and Stainless Steels

Los Alamos
NATIONAL LABORATORY

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Acronyms and Abbreviations

Acronym	Definition
AISI	American and Iron Steel Institute
LANL	Los Alamos National Laboratory
NNSA	National Nuclear Security Administration
HCL	Hydrochloric Acid
Ra	Surface Roughness
H_2SO_4	Sulfuric Acid
TEM	Transmission Electron Microscopy
NFT	Nuclear Filter Technology
ORI-2	Operational Readiness & Execution

1 The Influence of Surface Roughness on the Corrosion of Metal

Concerning the SAVY 4000 container LANL inspection acceptance of Ra (surface roughness) and its correlation to subsequent surface corrosion, a problem statement has been identified. The problem statement is as follows: When the 316L container has been subjected to accommodating materials producing high wattage, along with materials that eventually produce HCL (hydrochloric acid) will a higher or lower Ra on the inner surface of the SAVY 4000 container influence the rate or the amount of corrosion?

In an effort to address this concern, a literature review covering more fundamental studies that examine the influence of roughness on corrosion in metals are surveyed. The scope is kept relevant to the less dynamic storage conditions that a SAVY container may be exposed to.

1.1 How Does Ra influence Corrosion?

Surface roughness is often a result of a process that introduces a significant change to the material. In the case of SAVY containers, the deep drawing process introduces surface roughness dependent on both the process parameters and equipment used. The underlying phenomena and surface roughness itself play a key role in the corrosion rate of metals and has the potential to adversely influence the corrosion behavior over the course of component lifetimes. How surface roughness influences corrosion rates requires a knowledge of both electrochemical and geometric changes to the surface. Evgeny *et al.* highlight some of these [1]:

1) An increase in surface area from a rougher surface that gives rise to a proportionate increase in corrosion rates and a higher degree of wetting on a rougher surface.
2) An increase in the corrosion potential with an increase in roughness that stabilizes pitting corrosion. Does this, in turn, increase in the possibility of increasing sites that trap corrosion products and promote additional corrosion locally?
3) A change in sub-surface defects, such as dislocation content and residual stress, that can serve to influence corrosion rates.

The key to addressing the question of the effect of surface roughness on SAVY corrosion is understanding, with respect to the SAVY storage containers, which of these plays a dominant role in the corrosion behavior and where these arise from during the SAVY forming process. It is worth noting that discussion of the increased surface area is not included as the contribution is both intuitive and difficult to decouple from other effects.

1.2 Ra and Corrosion (Pitting) Potential

As a first order effect, surface roughness influences the pitting potential of a metal surface [1-3]. It was found for stainless steels that the precursor to stable pit growth, metastable pits, were a good qualitative indicator to the corrosion resistance of metals. Metastable pits are pits that form but may passivate before continued growth. Those that do not passivate and continue to pit become stable pits and are readily observed. Burstein and Pistorius studied the influence of surface roughness on metastable pit formation [2]. They found that a smoother AISI 304 wire electrode surface, generated by polishing with a P4000 grit, resulted in fewer metastable pits than a similar wire electrode polished with P1200 grit. This observations is supported by Sasaki and Burstein that also observed a decrease in the pitting potential with an increase in surface roughness, albeit with a larger range of coverage [3].

The authors explained that pit growth rates are controlled by diffusion of metal cations into the electrolyte solution. For stable pit growth to occur, some perforated cover for the pit that allows diffusion of metal cations and blocks passivation (i.e. Cr oxide formation) is needed. Without this, the pit simply passivates without the chance to continue growth. The authors suggest that the deeper pit sites, generated by the larger grit sizes, are generally less open to other species and thus require a lower current to maintain diffusion that leads to stable pit growth.

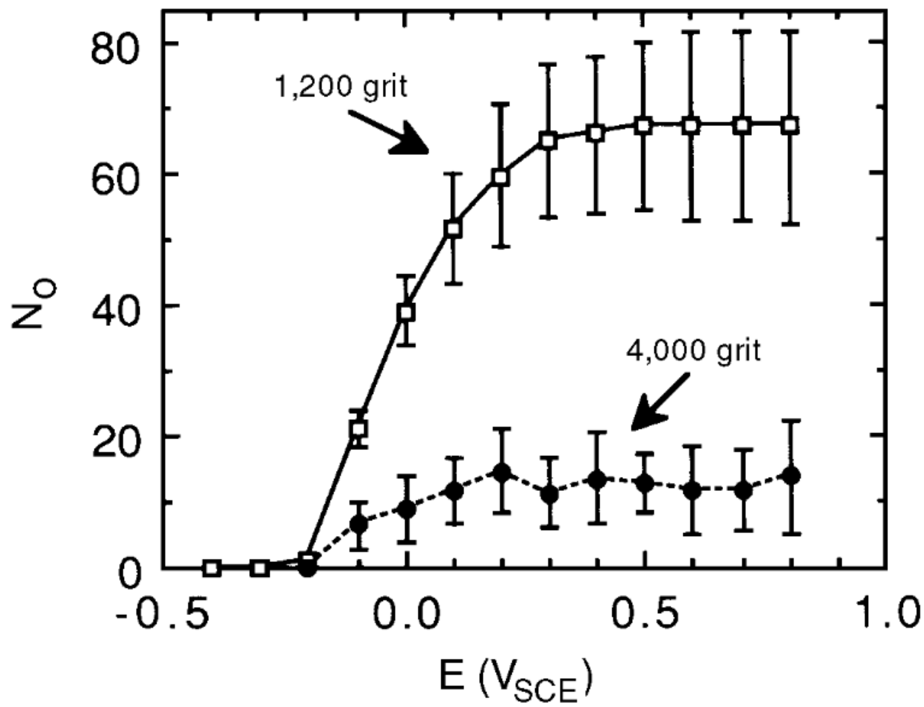

Figure 1. Number of metastable pit sites as a function of potential for AISI 304 wire in 0.8 M NaCl + 0.2 M HCl for each surface finish explored in [2].

This finding may have implications to corrosion behavior observed in deep SAVY pits. Those generated under storage environments tend not to be on a normal trajectory with respect to the inner wall surface but take a more tortured path through the wall. Although the previously mentioned studies focus on initial pit formation, the process may provide some insights into the path stable pit growth develops.

1.3 Ra and the Influence of Sub-surface defects and Residual Stress

Surface roughness arising from processing, such as grinding and polishing, is often accompanied by an increase in sub-surface defect density, such as dislocations, and residual stress. How these influence corrosion rates has been the subject of a number of studies, with many identifying practical routes, such as shot peening, to reducing corrosion [4-7]. It can be difficult to decouple the influence of surface roughness from the influence of defects and/or residual stress on corrosion rates unless special care is taken to remove one of these sources as a major contributor [8]. We provide coverage to two studies that show a clear influence of defect density (dislocation) and residual stress on the corrosion of steels.

Residual stresses have often been cited as a potential source of corrosion behavior modification during working group meetings. It was understood that in previous storage containers, such as the Hagan, this influence had been observed. SAVY containers, annealed after drawing, presumably do not have [much] residual stresses in the container walls and have a very limited residual stress in the weld regions near the electron beam-welded collar. Lingering questions were whether or not residual stresses (tension/compression) are detrimental and if all polishing processes generate the same stresses.

A study by Takakuwa and Soyama systematically show the influence that residual stresses play on the corrosion behavior of American and Iron Steel Institute (AISI) 316L stainless steel [9]. Residual stresses were initially introduced by production method via electro polishing (A), an angle grinder (36 grit, B), 240 grit (C), and 800 grit (D) final preparation steps. Residual stresses were measured using a conventional X-ray diffraction method.

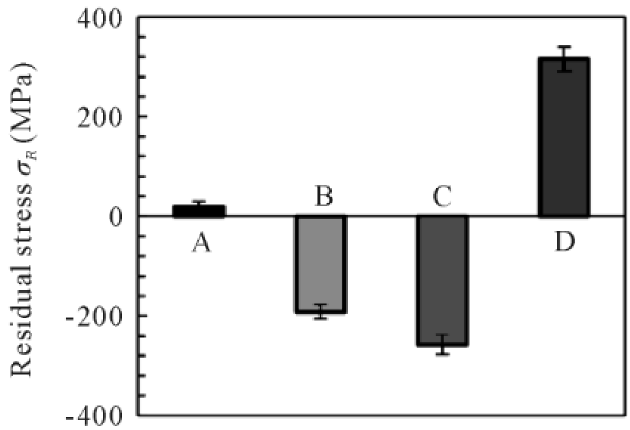

Figure 2. Residual stresses introduced by the four preparation methods [9].

Takakuwa and Soyama, acknowledging the influence of surface roughness on the corrosion behavior, sought to decouple surface roughness from residual stress. This was performed by using a cavitating jet on the back of the specimen surface that gradually increased the front surface (exposed to sulfuric acid (H_2SO_4)) compressive stress state. The results clearly showed that the critical current density for passivation decreased with increasing compressive residual stress. This, in turn, increases the corrosion resistance by facilitating passivation film formation.

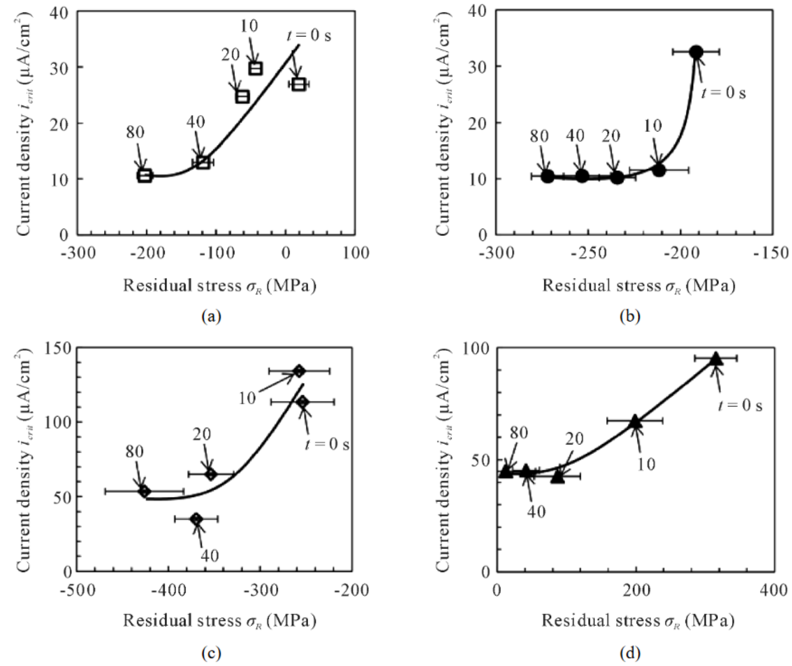

Figure 3. Critical current densities for passivation for each condition (A, B, C, D) with back surface removal to modify the front surface residual stress state [9].

It should be noted that residual stresses are not the only microstructural feature that can be accompanied by surface roughness. Microstructural defects, such as dislocations or grain boundaries, can be produced by the process that results in increased surface roughness. It is important to acknowledge this as a possible contributor to the corrosion behavior and explore how these defects influence corrosion rates.

Peguet et al. performed corrosion studies on both AISI 304 and AISI 430 stainless steels subject to cold rolling to understand the influence of this process on the corrosion rates [7]. The premise of this study was, in part, to determine which microstructural feature arising from the deformation of the material is a controlling factor in the corrosion behavior. The rolling and samples preparation process introduces both surface roughness and some amount of residual stress. Additionally, for austenitic stainless steels, a martensite phase can form under the strain caused by rolling and is also thought to play a role in the corrosion behavior.

To remove these effects, Peguet et al. stamped out disks and polished each to a diamond slurry of 3 um, followed by aging in air (it’s not explicitly stated what the conditions were). Performing a range of corrosion tests, including pitting potential and pitting transient measurements, the authors found that, on small scales, metastable pitting production (tied to stable pitting) was a maximum for an intermediate cold working step (20%). In the absence of microstructural data, this was in disagreement with the monotonic increase in pit propagation rate observed at the macroscale. Dislocations feature atoms with lower bonding energies to neighboring atoms than in a perfect crystal. This, in turn, enhances the dissolution of atoms and the overall corrosion rate. It appears that intuition would agree with the observations on the macroscale. Transmission Electron Microscopy (TEM) however, showed that the dislocation structure evolved from a planar forest of dislocations to a cellular dislocations structure, often a result of dynamic recovery or annealing. This suggested that dislocation structure can also influence corrosion rates, albeit locally.

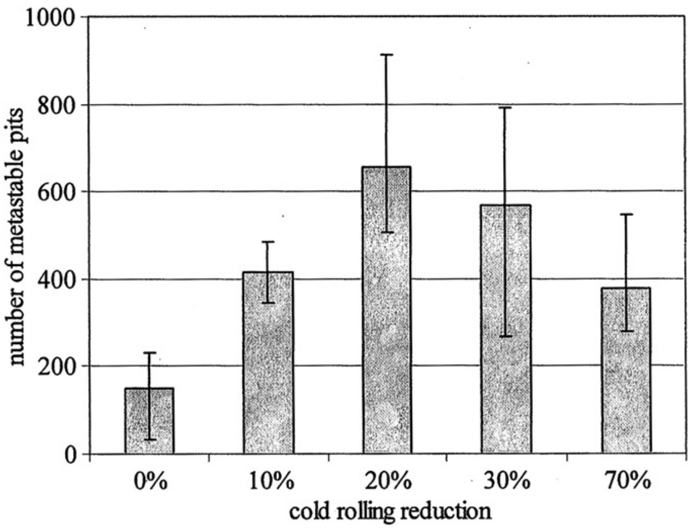

Figure 4. Number of metastable pits initiated during 24h on AISI 304 in a solution described in [7].

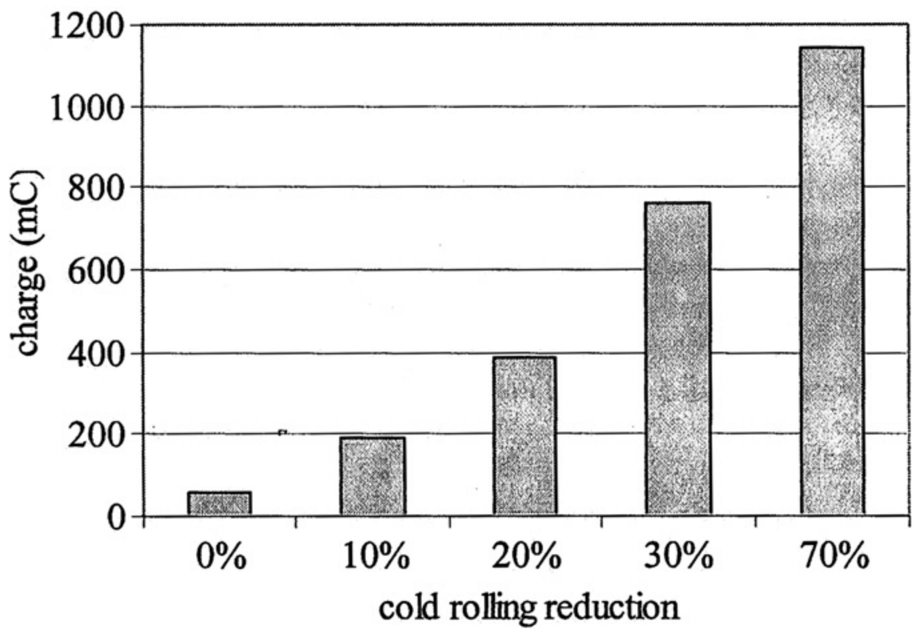

Figure 5. Total dissolved charge by metastable pitting during 24h for AISI 304 in a solution described in [7].

It is important to note that Peguet et al. mention the increase in potential difference between the inclusion and matrix caused by dislocation pileups can enhance pit initiation. It is well known that the SAVY container stock has inclusions present and these may play a significant role in localized corrosion behavior.

1.4 Ra and the Influence on Passivated Oxide Layer Structure

SAVY containers are routinely subject to passivation prior to shipment as a means of improving the inert nature of the container. There are a few studies that examine the influence of surface roughness on the performance of passivated stainless steels. We showcase one particular study performed by Shahryari et al [10]. In this study, the authors prepared steels with varying surface roughness values using conventional grinding paper. One set of specimens was passivated using a cyclic potentiodynamic passivation (CPP) technique while other was subject to no passivation.

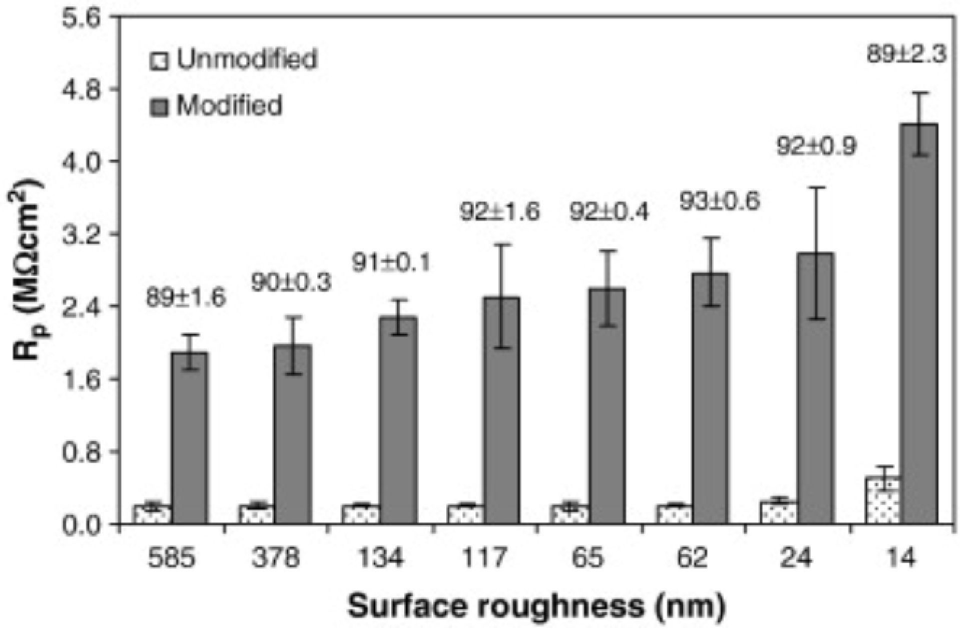

Figure 6. Polarization resistance of 316L (vacuum melted) with and without CPP at different surface roughness values.

The results clearly showed an improved corrosion performance for the passivated stainless steel. Both 316 specimens with and without CPP show a larger polarization resistance (corrosion resistance) with decreasing surface roughness. The results from this study suggest that care should be taken to produce as smooth of a surface finish as possible to improve general corrosion performance.

It is important to note that this trend applies only to oxide layers grown through CPP. The techniques used at NFT may be different and result in a different dependence on surface roughness. Furthermore, little information is known, at this time, about the relative corrosion performance with the passivation technique used at NFT and whether or not it provides significant improvements to the corrosion performance, regardless of surface roughness.

1.5 Low Ra on Various Stainless Steels Improves Corrosion Resistance

Of the literature currently researched, several sources were found to assist in reaching a clear consensus of the solution to the aforementioned problem statement. In one study and subsequent research completed by its authors, studies have shown that lowering the Ra on different types of stainless steels improves corrosion resistance [10]. In that study, the high contact angle of a water droplet on a metal surface is considered to have a high wettability and is said to be hydrophobic – low affinity for water, which was found to be associated with lower Ra(s) as shown below [11]. Ra acts as a porous medium for the liquid as represented by the Ra grooves in the figure below.

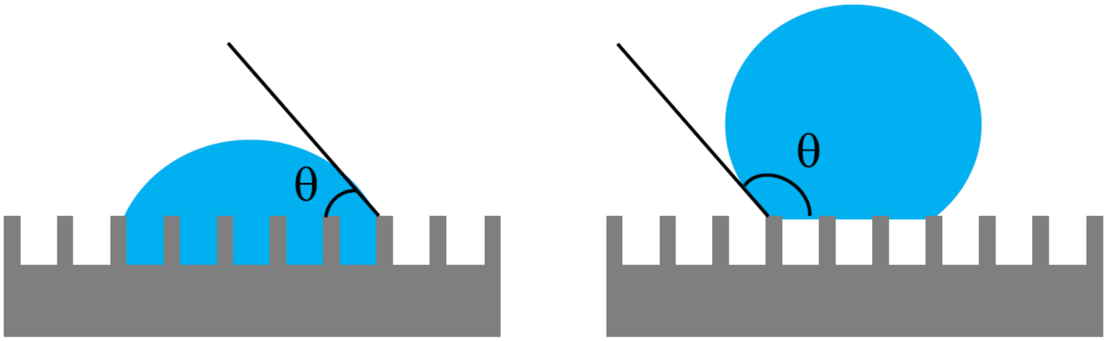

Figure 6. Illustration of Wetting on rough surfaces (a) Wenzel State (b) Cassie-Baxter state described in [10].

It has been found that Ra is a major influence on pitting corrosion, and that a smoother surface finish reduced the incidence of metastable pitting of stainless steel [12]. In another study, it was stated that observed behavior agreed with a microscopic model that attributes the initiation of pitting corrosion to the production and persistence of gradients of acidity and electrode potential on the scale of the Ra [13].

2 Summary of Findings

From our initial thought process on this problem statement and from the several sources in the literature concerning corrosion and Ra, we have to side on the premise that yes, Ra does affect the rate and/or amount of corrosion. From the literary research the below key points summarizes our findings.

1. Rougher surfaces exhibit less corrosion resistance owing to more favorable conditions for stable pit growth.
2. Surface roughness is often accompanied by microstructural changes that can influence corrosion rates.
    a. An increase in the tensile residual stress increases the corrosion rate. Compressive residual stresses serve to lower corrosion rates by a lowering of the critical passivation current density.
    b. An increase in the near surface dislocation content increases the corrosion rate.
3. Surface roughness influences corrosion performance of passivating layers with an increase in performance tied to a decreasing surface roughness.

The higher the Ra, the rougher the surface, the more corrosion. Conversely, the lower the Ra or smoother the surface, the less corrosion. However, we believe that in order to see differences in the corrosion, the Ra may have to be lowered significantly like for e.g. from 63 to 32 or 63 to 16. LANL expectations of the SAVY(s) manufacturer, Nuclear Filter Technology (NFT), to provide low Ra(s) for the internal walls of the SAVY may be unrealistic. More research may be necessary to explore other ways to reduce the effect of corrosion on 316L such as surface chemistry.

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